Non-Linear Creep Behaviour of Polycarbonate

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The non-linear viscoelastic behaviour of polymers raises persistent challenges to accurate physical and mathematical modelling. Real long-term predictive capability is the most sought-after objective in the researchers' endeavours to unravel the seemingly coupled effects of time, temperature and mechanical stress on the creep compliance of materials. Most recent work concentrated on recognizably useful but semi-empirical, lumped-parameter, modelling approaches that extended and rationalized much earlier experiments and formulations.

We adopted a (non-simulative, mainly analytical) truly dynamic molecular modelling approach to general compliance and relaxation behaviour, whereby actual elementary, molecular scale, process-relevant frequencies are derived by adequate (as simple as possible) kinetic formulation. These elementary processes follow an almost exactly Arrhenius behaviour, with a range of activation enthalpies, but their relative contribution to the overall macroscopic behaviour of the materials is adequately quantified, to account for the materials' retardation time spectra and final non-Arrhenius and stretched-exponential behaviour. Examples of such elementary molecular scale processes might be, in simple linear polymer chains, the crankshaft-type main chain motions.

The analysis of the experimental data for the polycarbonate (PC) show that there is a minimum retardation time (τ_1), corresponding to the smallest contributing clusters (n = 1) in the least constrained local environment within the structure. For an infinite number of log-normally distributed elementary retardation times, it may be show that the creep compliance may be formulated as

$$D(t) = D_0 + (D_\infty - D_0)\frac{erf\left[b\ln\left(\frac{\tau^*}{\tau_1}\right)\right] + erf\left[b\ln\left(\frac{t}{\tau^*}\right)\right]}{erf\left[b\ln\left(\frac{\tau^*}{\tau_1}\right)\right] + 1} \qquad (1)$$

The effects of temperature and applied stress on the optimised values of b, τ_1, τ^*, D_0 and D_∞ (actual values and physical significance) that best fit the experimental creep strain values are discussed. Figure 1 show the results of fitting Eq. (1) to the experimental creep compliance curves obtained for PC at 40 ºC.

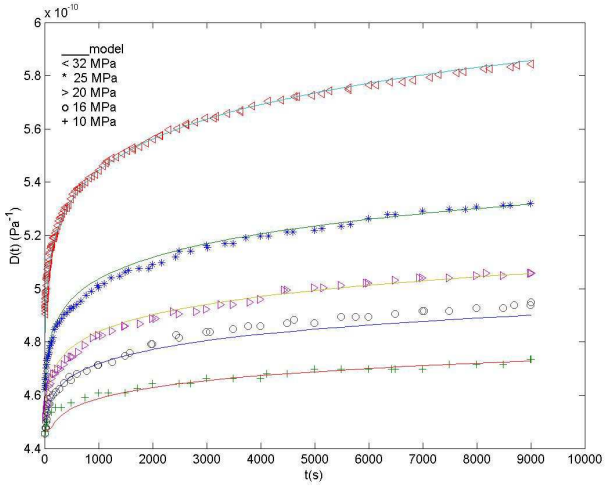

Figure 1. Model fitting (Eq.1) of the Creep Compliance of PC at 40 ºC.